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Comparison of Pr-doped Ca 122 and Ca 112 Pnictides by Low-field Microwave Absorption Spectroscopy

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ABSTRACT

Praseodymium doped CaFe_2As_2 (122 structure) and CaFeAs_2 (112 structure) are characterized by modulated Low Magnetic Field Microwave Absorption (LFMA) spectroscopy. In both (Pr,Ca)122 and (Pr,Ca)112 structures, a strong hysteretic LFMA is found, with a T_c^H of ~ 30 K and ~ 26 K, respectively. However, in (Pr,Ca)122, measurements also show an unusual Narrow Peak (NP) LFMA signal appearing at higher temperatures, above the lower T_c^H superconducting state until a T_c^{NP} of 49 K. We associate this NP LFMA with interfacial superconductivity, which has been found previously by highly anisotropic magnetization measurements. Furthermore, the absence of NP in (Pr,Ca)112 correlates with the absence of an interfacial phase. These results give useful information about the microwave signature of interfacial superconductivity present in the (Pr,Ca)122 system, and may form a roadmap towards a stabilized high temperature superconducting phase in pnictides.

INTRODUCTION

Among the recently discovered class of doped pnictide superconductors, the so-called 122 systems ($M\text{Fe}_2\text{As}_2$ with M an element of valency 1 or 2 [1]) have attracted significant interest [1-5]. In particular, superconductivity in the studied Ca122 pnictide has been found to be significantly different from a seemingly similar system, Ca112 [6,7]. The maximum T_c in reported 122 systems is only 38 K [1,5]; however, a recent independent observation of a high temperature phase appearing between $T_{c1} = 21$ K and $T_{c2} = 49$ K in Pr-doped Ca122 [2] opens a new perspective to search for even higher critical temperatures. The exceedingly small volume fraction, ease of suppression by magnetic field, absence of the characteristic specific heat anomaly of superconductivity [2], and the high magnetic anisotropy [8] imply that the high temperature phase is formed from interfaces present in the crystal. This “interfacial superconductivity” may in the future be improved or stabilized [9], leading the way for high temperature superconducting (HTSC) pnictides. In this paper, we report results of modulated low magnetic field microwave absorption (LFMA), a powerful technique in examining the microscopic superconducting behavior of each system. The (Pr,Ca)122 system exhibits an unusual signal, with a very narrow absorption peak (NP) centered on zero field, between $T_c^{NP} = 49$ K and a transition to a hysteretic signal below around $T_c^H = 30$ K. This type of signal is not observed in the (Pr,Ca)112 pnictides.

EXPERIMENTAL DETAILS

(Pr,Ca)122 and (Pr,Ca)112 single crystals were grown from the self-flux method, as described

previously by Lv, et al [2]. For LFMA measurements, the 122 crystals were affixed to a quartz capillary tube and sealed inside a 4mm EPR ampoule (Wilma LabGlass) at $\sim 10^{-6}$ torr. The crystals could be oriented relative to the measurement apparatus. LFMA was performed inside a standard electron paramagnetic resonance (EPR) spectrometer (Bruker EMX), fitted with an X-band (9.8 GHz) microwave source and TE₁₀₂ cavity. Temperature control is achieved by a ColdEdge cryogen-free cryostat, capable of reaching $T = 4$ K under flow of helium transfer gas.

The fields inside of the cavity are shown in Figure 1. The sample is exposed to three magnetic fields: in the x direction, a slowly sweeping (2.5 G/s) magnetic field H_0 (provided by a pair of Helmholtz Coils, generally ramping between -50 and $+50$ G) and a sinusoidally modulating field of intensity on the order of 1 G and frequency of 100 MHz (provided by modulation coils within the cavity); and in the z direction, a magnetic field from the standing microwaves [10]. The static and modulation fields form vortices or fluxons within a Type-II superconducting material, which are oscillated about their pinning centers by the microwave currents [11]. This absorbs a portion of the microwave energy, resulting in a signal [12].

Magnetization measurements are performed in a Quantum Design MPMS.

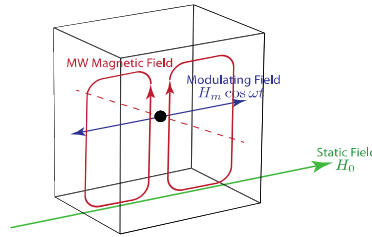


Figure 1: Orientation of magnetic fields inside the TE₁₀₂ resonant cavity of the EPR device, which is used for LFMA measurements. The sample location is denoted by a black circle.

RESULTS AND DISCUSSION

The typical LFMA spectra observed for (Pr,Ca)122, with $H_{mw} \parallel ab$ planes, upon zero field cooling of a sample to 4 K and increasing T slowly are shown in Figure 2(a) and Figure 2(b), where $H_0 \parallel ab$ planes or c axis of the crystal, respectively. An increased modulation field can bring out smaller features with less noise, at the expense of possible loss of detail. An increased microwave power increases the screening currents responsible for oscillations about the fluxon's pinning centers. [11,13]

LFMA shows two distinct shapes in temperature regions $T < T_c^H = 30$ K and $T_c^H < T < T_c^{NP} = 49$ K. Below T_c^H , we see relatively typical LFMA signals for a superconductor with both flux trapping and Josephson decoupling [14]. Intensity and hysteresis width decrease with increasing temperature. Between T_c^H and T_c^{NP} the signal changes dramatically. Hysteresis nearly vanishes, but a strong, narrow zero field peak, or NP, appears similar to that arising from Josephson junction (JJ) decoupling found earlier in HTSC [12]. The field width of this signal is only ~ 2 G. It is worth noting that the width of the NP is also observed in $M(H)$ curves, in the $T > T_c^H$ regime, as the value of H_{c1} , the minimum of the characteristic “butterfly” curve of superconductivity (discussed below).

We may compare these results with those in Figure 2(c,d), which differ only by a rotation of the sample about the z axis, that is, where $H_0 \parallel c$ axis of the crystal, but still with $H_{mw} \parallel ab$ planes. Qualitatively the behavior is similar, with a transition from the hysteretic LFMA of the low temperature phase to the NP signal of the high temperature phase. However, the signal

intensity is higher (which may be partially due to the demagnetizing factor), and the NP signal appears at a lower temperature than in the previous case. A similarly large anisotropy, supporting the hypothesis of interfaces, is also observed in magnetic measurements [15].

The temperature dependence of intensity, as shown in Figure 3, reveals just how dramatically the NP signal appears in measurements. The high temperature phase manifests as a sharp increase in peak-to-peak signal intensity, without a corresponding increase in the hysteretic signal. We associate this NP signal with the “interfacial” phase. We also note that while the low temperature phase was only observed below $T_{cl} = 21$ K previously [2], we observe hysteresis until $T_c^H = 30$ K. We attribute this to the high sensitivity of the LFMA technique.

We may further analyze this signal by fitting the temperature dependence of LFMA intensity to the Josephson junction model of Nebendahl [16]. The model states that LFMA intensity, as a function of temperature, should fit an equation of the form:

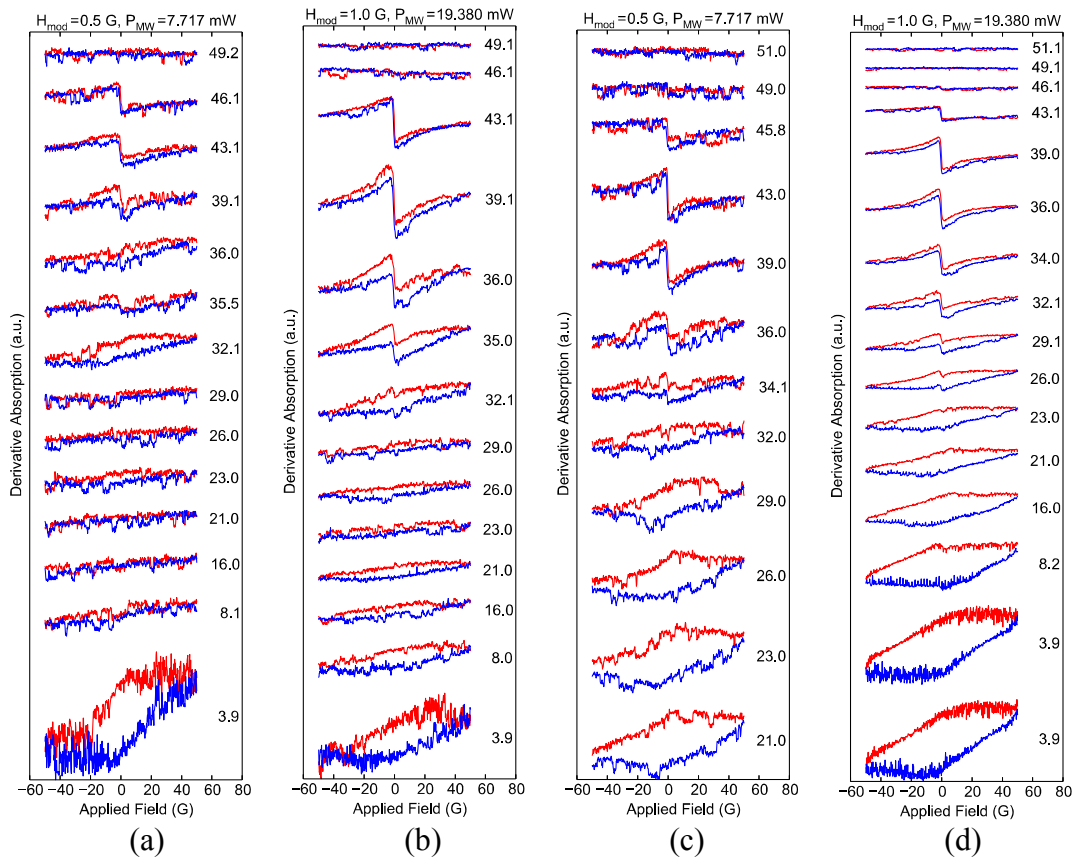


Figure 2: LFMA spectra of (Pr,Ca)122 sample, with the orientation fixed such that $H_{mw} \parallel ab$ planes; (a,b) $H_0 \parallel ab$ planes of the crystal, and (c,d) $H_0 \parallel c$ axis of the crystal. Each stack of curves represents a specific combination of microwave power and sinusoidal modulation field amplitude. The number to the right of each curve is the temperature of measurement in Kelvin. A dramatic transition is clearly seen in the vicinity of 30 K. Below that transition, the LFMA exhibits a purely hysteretic signal, from trapped flux within the superconducting state, while above the transition, a narrow field width absorption peak (NP) appears in the signal, with the absence of any hysteresis, and vanishes at ~ 49 K, the temperature observed by other methods as the onset of “interfacial superconductivity.” All curves (a-d) have a clockwise hysteresis.

$$I_{pp}(T) = a_0 \frac{1}{(1 + \eta_0 (1 - T/T_c)^{2\alpha})^{3/2}} \left(1 - \frac{T}{T_c}\right)^{2\alpha} \quad (1)$$

where a_0 , η_0 , T_c , and α are determined from curve fitting. The α parameter is particularly important, as it indicates the type of Josephson junctions (JJ) present: $\alpha=1$ for SIS-type junctions (superconductor-insulator-superconductor), $\alpha=2$ for SNS-type junctions (superconductor-normal metal-superconductor).

Assuming that the NP feature is due to JJs, we can fit the $H \parallel ab$ curve with $T_c = 48.3 \pm 0.6$ K, $\alpha = 2$, $\eta_0 = 1.23 \times 10^3$, and $a_0 = 5.04 \times 10^5$. ($H \parallel c$ gives similar fitting coefficients.) Not only is the predicted T_c^H of 49 K confirmed, but also the behavior of the high temperature superconducting phase is determined to be from SNS-type Josephson Junctions.

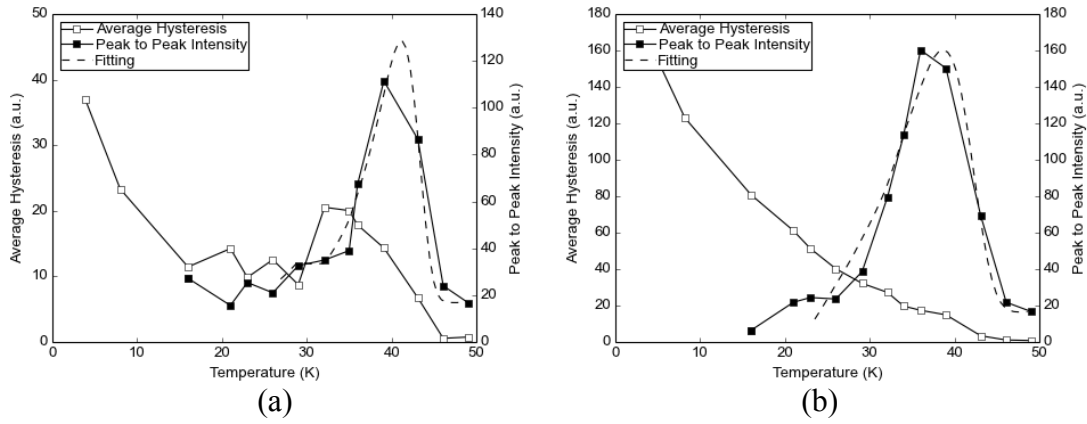


Figure 3: Intensity of hysteresis (open squares) and NP (filled squares) versus temperature, where $H_{\text{mod}} = 1$ G, $P_{mw} = 19.43$ mW, for orientation (a) $H_0 \parallel ab$, and (b) $H_0 \parallel c$. The dashed line is the fit to the model of Nebendahl (Eq. 1). The appearance of the NP is clearly visible as a spike in the total intensity curve which does not appear in the hysteresis intensity dependence.

The (Pr,Ca)112 system has a substantially different behavior in LFMA measurements. Typical LFMA spectra observed upon zero field cooling of a sample to 4 K and increasing T slowly are shown in Figure 4. We observe two noticeable differences between the 112 and 122 systems. Firstly, the (Pr,Ca)112 results show no indication of a higher temperature NP-type signal, only the hysteretic signal of the low temperature phase below 23 K. Figure 5(a) shows the falloff of intensity with temperature, and no additional NP-type signal. (This behavior is correlated by magnetization in Figure 5(b); the zero field cooled and field cooled curves split above 40 K for (Pr,Ca)122, but remain together until close to 20 K for the (Pr,Ca)112. We also note the correlation between the width of NP and H_{c1} (inset) as discussed previously.) Secondly, the strength of the signal decreases dramatically with increasing microwave power (see inset of Figure 5(a)), an effect which was not observed in the 122 case. In fact, the strength of the LFMA spectra for (Pr,Ca)122 increases with microwave power.

CONCLUSIONS

Low-field microwave absorption is a powerful and highly sensitive technique for characterizing complex, multiphase superconducting materials such as the (Pr,Ca)122 and (Pr,Ca)112 single crystalline systems. We have demonstrated the ability to detect small volume

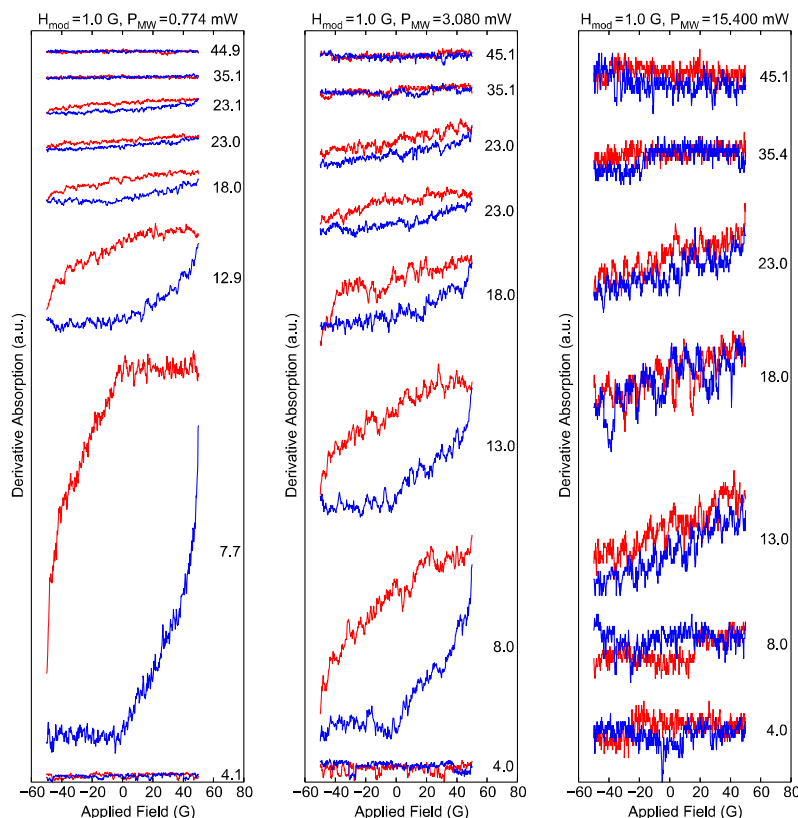


Figure 4: LFMA scans of (Pr,Ca)112 sample for various microwave powers, and hence magnetic fields. (All scans performed with modulation amplitude of 1 G.) Increased microwave power causes a rapid decrease in LFMA hysteresis. All curves have a clockwise hysteresis.

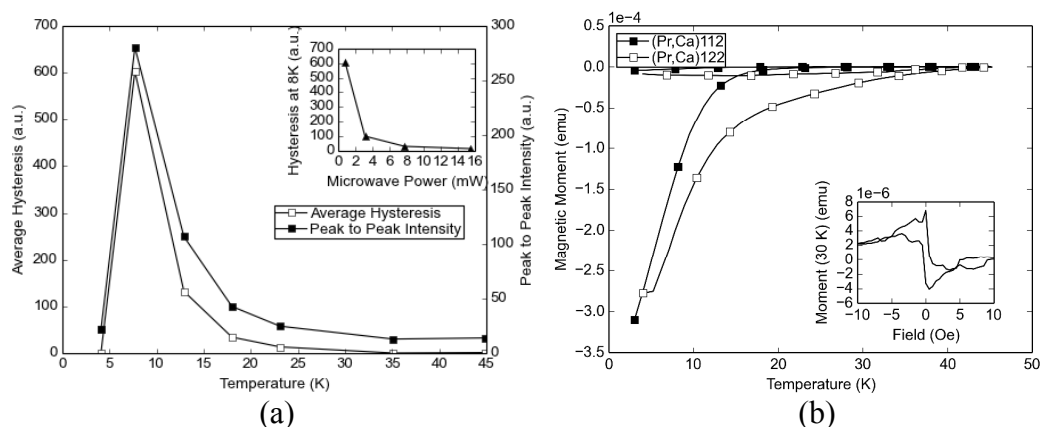


Figure 5: (a) Main plot: dependence of hysteresis (open) and peak-to-peak (filled) intensity on temperature for (Pr,Ca)112 with $H_{\text{mod}} = 1$ G, $P_{\text{mw}} = 0.774$ mW. We note the absence of any high temperature phase appearing in either curve. The offset between curves is due to the electronic background noise. Inset: Strength of hysteresis at 8K, with $H_{\text{mod}} = 1$ G, as a function of applied microwave power. (b) Main plot: Zero field cooled (lower) and field cooled (upper) magnetization data with an applied field of 10 Oe, comparing (Pr,Ca)122 with (Pr,Ca)112. We note that the 122 pnictide ZFC data deviates from the FC data above 40 K, while the 112 pnictide ZFC does not deviate from FC until near 20 K. Inset: $M(H)$ measurement for (Pr,Ca)122 at 30 K, showing the same small H_{c1} as LFMA. (Counter-clockwise hysteresis.)

fraction [2] phases by hysteretic LFMA in both systems, and exceedingly small volume fraction by NP LFMA, with higher T_c^{NP} , which only appears in (Pr,Ca)122. We can correlate the appearance of the NP with interfacial superconductivity previously reported. Additional analysis will be required in order to fully understand the complicated nature of multiphase superconductivity in these electronically doped Ca122 and Ca112 pnictides.

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